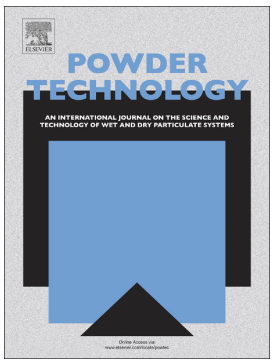


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PII: S0032-5910(23)00100-6

DOI: https://doi.org/10.1016/j.powtec.2023.118316

Reference: PTEC 118316

To appear in: *Powder Technology*

Received date: 10 December 2022

Revised date: 11 January 2023

Accepted date: 31 January 2023

Please cite this article as: F.E. Vasile, D.A. Archaina, J. Jiménez-Guzmán, et al., Prosopis alba exudate gum as new carrier agent for obtaining powdered Hibiscus sabdariffa aqueous extracts by spray drying, *Powder Technology* (2023), https://doi.org/10.1016/j.powtec.2023.118316

# *Prosopis alba* exudate gum as new carrier agent for obtaining powdered *Hibiscus sabdariffa* aqueous extracts by spray drying

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**ABSTRACT**

The objective of this work was to study the functional behavior of *Prosopis alba* exudate gum (G) as component of powders rich in antioxidant pigments. For this purpose, the morphological, structural and chromatic characteristics as well as physical, biological and stability properties of spray dried aqueous extracts of *Hibiscus sabdariffa* were evaluated for different ratios of maltodextrin and G, and opportunely compared with gum arabic (GA). Powders containing G showed low moisture (4.9% ds.), low water activity ($a_w$<0.3), high solubility (>95 %) and small (~25μm) spherical-shaped particles, with a glass transition temperature ($T_g$ = 52 °C), similar to those containing GA. Phenolic compounds, naturally present in G, improved the storage stability of Hibiscus extract pigments contributing to the preservation of the antioxidant properties. Results encourage to consider *Prosopis alba* exudate gum as a promising alternative to gum arabic, with the subsequent benefits stemmed from adding value to an underutilized resource.

**Key words**

- *Prosopis alba*
- *Hibiscus sabdariffa*
- Spray drying
- Anthocyanin
- Antioxidant

**Abbreviations:** F-HE (freshly prepared hibiscus aqueous extract), HE (spray dried hibiscus aqueous extract), G (*Prosopis alba* exudate gum), GA (gum arabic), MD (maltodextrin), HEMD (Hibiscus extract with maltodextrin), MDG (Hibiscus extract with maltodextrin and *P. alba* exudate gum), MDGA (Hibiscus extract with maltodextrin and gum arabic), $T_g$ (glass transition temperature),RH (relative humidity), GAE (gallic acid equivalent), ABTS (2,2 azino-bis [3-ethylbenzo-thiazoline-6-sulfonic acid]), TE (Trolox equivalent), FRAP (ferric reducing ability), TPTZ (2,4,6-Tris[2-pyridyl]-s-triazine), VCE (vitamin C equivalent), C (croma), h (hue), ΔE (Total color difference), CIEL*a*b* (Commission Internationale de l'Eclairage color space), FT-IR (Fourier-transform infrared spectroscopy), ATR (attenuated total reflectance), BI (browning index), PC (polymeric color), ANOVA (analysis of variance).

## 1. Introduction

*Prosopis* trees are widely dispersed in the arid and semi-arid regions of South America. Like most leguminous plants, under environmental stress conditions, *Prosopis* spp. exudes a water-soluble gum. Particularly, *P. alba* was recently reported as a promising source of hydrocolloids. The *P. alba* exudate gum shows interesting functional properties, as emulsifying [1], and antioxidant [2], which turns G into a potential food additive. However, there still exist some gaps in the knowledge of the technological behavior of G, which hinders its utilization on an industrial scale.

Exudate gums are widely used in the food and pharmaceutical industries. Particularly, gum arabic (GA) has been highly preferred due to its high water solubility, low viscosity -even at high concentrations- and excellent emulsifying characteristics [3, 4]. Frequently, GA is combined with maltodextrin (MD) in aqueous dispersions to build stable, free-flowing, and easily dispersible powders by spray drying, [5, 6]. Despite the usefulness of GA, there has been an increasingly growing demand for new, low-cost, and locally available alternatives. Thereby, several unconventional exudate gums have been recently explored for many applications and processes such as spray drying [7-9].

*Hibiscus sabdariffa* L. is widely grown in tropical and subtropical areas of both hemispheres [10, 11]. In many countries, Hibiscus calyxes, also known as “Roselle” or “Jamaica”, are used to prepare slightly astringent and sour aqueous infusions, which are consumed as hot or cold beverages [12]. The aqueous extracts of *H. sabdariffa* exhibit interesting antibacterial, antioxidant, nephro- and hepatic-protective, renal, diuretic, antidiabetic and antihypertensive effects, as well as benefits on lipid metabolism (anti-cholesterol), among others [6, 9, 13]. Most of these health-beneficial properties were attributed to the presence of phenolic and organic acids, flavonoids, and anthocyanins, which also impart unique chromatic properties [9]. Due to this facts, Hibiscus extract constitutes a very interesting source of nutraceutical compounds, providing health benefits along with attractive red to violet colors [11]. However, preserving the bioactivity and functionality of Hibiscus antioxidant pigments requires addressing specific strategies to achieve stability against adverse environmental conditions (light, oxygen, moisture, and high temperatures) [14].

Dehydration by spray drying is still the best option to stabilize biologically active plant substances, due to its simplicity [15], low cost, relative speed of this process [10], one-step continuous process, and easy scaling [16]. As stated by Shamaei et al. [17], producing powders with high quality, low water activity, and good shelf-life by spray drying implies four main steps (i) selecting drying agents, (ii)

preparing emulsion or slurries containing oils, antioxidants, bioactive phenolics, etc., (iii) feeding the mixture into a drying chamber through a nozzle, and (iv) collect and air-tightening the powder obtained. In most cases the addition of drying agents or *carriers* is necessary to improve the physical and chemical properties of the dehydrated substances [6, 18]. A wide variety of biopolymers such as proteins (whey proteins, caseins, etc.), carbohydrates (starch, maltodextrin, among others) and gums has been used as carriers [4]. As expected, the nature of the drying agents and composition of the spray dryer feed mixture strongly influence the quality of the obtained particles and determine the final products application [3, 19]. To contextualize the originality of present work, most of the recent studies related to obtaining functional ingredients rich in antioxidants and pigments from *Hibiscus sabdariffa* extracts through drying or gelation are summarized in Table 1. With this background, the aim of this work was to assess the functional behavior of *P. alba* exudate gum as pigment and bioactive ingredient in spray dried powders of aqueous Hibiscus sabdariffa extract. Physical, morphological, biological, functional, and stability aspects of the obtained powders were explored for different gum-maltodextrin ratios and eventually compared with gum arabic.

(Table 1)

## 2. Materials and methods

### 2.1. Materials

Dehydrated *Hibiscus sabdariffa* calyxes were commercially acquired in a market in Mexico City. *Hibiscus sabdariffa* was chosen because of its high concentration of anthocyanins among other sensitive bioactive compounds [10]. *Prosopis alba* exudate gum (G) from the northeast region of Argentina was purified and freeze-dried as described previously [1]. Gum arabic (GA) from *Acacia* trees was purchased from Sigma-Aldrich Corporation (St Louis, MO, USA), and Maltodextrin (DE=20) was acquired from CPI Ingredients S.A. de C.V. (State of México, México). Folin–Ciocalteu reagent, 2,2-azinobis-3-ethylbenzotiazoline-6-sulfonic acid (ABTS), 2,4,6-Tris(2-pyridyl)-s-triazine (TPTZ), Gallic acid, Trolox and ascorbic acid (Vitamin C), were purchased from Sigma-Aldrich Corporation (St Louis, MO, USA). All the other reactants were of analytical degree, commercially available, and used as received.

### 2.2. Preparation of Hibiscus extract and feeding mixtures

Hibiscus extract (F-HE) was prepared according to the optimized extraction method proposed by Sindi, Marshall, and Morgan [20]. Briefly, Hibiscus calyxes were subjected to decoction in boiling water in a 1:10 ratio for 10 min. After cooling down, the extract was filtered through a Whatman No.1 filter with help of a Buchner funnel. For each drying batch, 50 g of a carrier blend were dispersed in 2000 mL of recently prepared extracts (6.4 % ds.) and subjected to mild stirring at least for 60 min under to ease the complete dispersion of hydrocolloids.

Different carrier blends with maltodextrin (MD) and *Prosopis alba* exudate gum (G) were prepared: 100% MD (HEMD), 95% MD + 5% G (MDG1), 85% MD + 15% G (MDG2), 70% MD + 30% G (MDG3). Additionally, a formulation containing gum arabic (GA) at intermediate gum concentration was prepared: 85% MD + 15% GA (MDGA2). In all formulations, the final solid content was 8.8 g solids/100 mL. Feeding mixtures were maintained overnight at room temperature and protected from light, to allow the biopolymer hydration before spray drying. For comparative purposes, a Hibiscus aqueous extract with no carriers (HE) was dried as a blank formulation.

### 2.3. Spray drying

A pilot-scale spray dryer (Mobile Minor 2000, GEA, Denmark) was used. Feeding suspensions were introduced using a peristaltic pump (Watson Marlow 520S, USA) at a feed flow rate of 12 rpm. The inlet temperature was 180 ± 3 °C, and the outlet air temperature was 80 ± 1 °C. Suspensions were pulverized in a co-current two-fluid nozzle system setting the air pressure at 0.8 Bar. The collection of coarse and fine particles was performed in a cyclone equipped with a collection vessel. Samples were recovered in opaque and hermetical bags and stored in a desiccator at 25 °C until further analysis.

### 2.4. Examination of physicochemical properties of powders

#### 2.4.1. Moisture and water activity

The moisture of powders was gravimetrically determined at 105 °C until constant weight. The moisture was expressed as g of water/100 g of dry solids (% ds.). The water activity ($a_w$) of powders was measured using a water activity meter AquaLab (Pullman WA, USA) at 25 °C.

#### 2.4.2. Hygroscopicity, wettability and solubility

Hygroscopicity was evaluated according to do Valle Calomeni et al. [21] with some modifications. Briefly, 0.1 g of powder was placed in glass vials and incubated for 7 days under controlled conditions (relative humidity of 75 % and 25 ± 1 °C). The hygroscopicity of samples was determined by measuring the water adsorbed after 7 days. Results were expressed as g of adsorbed water/ 100 g of dry solids (% ds.).

The wettability of samples was determined based on estimating the time (s) required to immerse 1 g of powder on a surface of 400 mL of distilled water at 25 °C [19].

The solubility was evaluated according to the method proposed by do Valle Calomeni et al. [21] with slight modifications. About 0.5 g of powder was added to 50 mL of distilled water and stirred by a magnetic stirrer for 5 min at 25 °C. Then, the mixture was centrifuged at 3500 g for 5 min. An aliquot of 25 mL of the supernatant was dried at 105 °C, until constant weight. Then, the solubility (%) was calculated using Eq. (1):

$$Solubility(\%) = (A/B) * 100\% \quad (1)$$

Where A is the weight of dry solids in supernatant and B is the weight of the dry powder (g) used to prepare the solution.

2.4.3. Glass transition temperature

The glass transition temperature ($T_g$) of samples was determined by DSC (differential scanning calorimetry) (Mettler TA 4000, Columbus, Ohio, USA). About 5 mg of sample was heated on a 40 µL aluminium pan (Mettler) from - 80 to 100 ºC (dynamic method) at a rate of 10 ºC $min^{-1}$. An empty pan was used as a reference. $T_g$ was recorded as the onset temperature at which a change in baseline of the curve of heat flow versus temperature was determined. $T_g$ was explored in samples equilibrated at 11 % RH.

2.4.4. Particle morphology and size distribution analysis

Morphology of microparticles was evaluated with a scanning electron microscope LSM 710 NLO (Carl Zeiss, Germany) equipped with a field emission gun, detector InLes and a third generation column GEMINI®. Powder samples were mounted on a specimen holder using double adhesive tape and coated with a thin layer of gold under vacuum. Images were collected at 500x, 1000x and 2500x. The mean

particle size (evaluated as particle diameter) and shape parameters of powder were performed by laser diffraction in a CILAS 1090 ExpertShape (La Source, Orleans Cedex, France). The results of the analysis of 1200–2000 particles from each sample were expressed as the punctual estimation of the measurements using the method of moments with an estimation of the geometric distribution.

2.5. Bioactive compounds, antioxidant capacity, and chromatic properties

About 100 to 150 mg of spray dried samples were dispersed in 10 mL of double distilled water and were left standing at least 15 min at room temperature to complete the hydration and dissolution and kept in dark until analysis.

Total phenolic content was evaluated using the Folin-Cicolteau method as described by Shen et al. [22]. Phenolic compounds were quantified as gallic acid equivalent GAE/mg HE ds., using the calibration curve y=7.785x ($R^2$=0.99).

Total monomeric anthocyanins were measured through the pH differential method, according to Giusti and Wrolstad [23]. Pigment content was determined from absorbance measured at $\lambda_{max}$= 520 nm, and using the molecular weight and molar extinction coefficient of cyanidin-3-glucoside (MW=449.2 g/mol; ε=26,900 L/mol·cm), and water as blank. Then, the results were expressed as mg cyanidin-3-*O*-glucoside equivalent/mg HE ds.

The antioxidant capacity was analyzed in terms of $ABTS^{\cdot+}$ radical scavenging activity [24]. For this purpose, 7 mM of $ABTS^{\cdot+}$ solution was prepared by dissolving 2,2 azino-bis (3-ethylbenzo-thiazoline-6-sulfonic acid), ($ABTS^{\cdot+}$) in 2.5 mM potassium persulfate and kept overnight in the darkness at room temperature. The $ABTS^{\cdot+}$ radical solution was mixed up with double distilled water to obtain an absorbance of 0.70 ± 0.2 at 754 nm. After each trial, 3 mL of $ABTS^{\cdot+}$solution was added to the glass cell and the absorbance ($A_0$) was measured. Then, 20 µL of sample solutions were added. Absorbance was monitored through time and the kinetic profile was fitted to one phase decay model to obtain the absorbance at equilibrium ($A_f$). Then, the percentage of inhibition was calculated using Eq. (2):

$$\%I = \frac{(A_0 - A_f)}{A_0} * 100\% \quad (2)$$

The antioxidant capacity was calculated using a Trolox standard curve $y=1.09\cdot10^6x$ ($R^2$=0.99). Where *x* is the concentration of Trolox (mmol/mL). The results were expressed as mmol Trolox equivalent TE/100 g HE ds.

On the other hand, the ferric reducing ability of samples was assessed. Briefly, 3 ml of freshly prepared FRAP reagent (300 mM acetate buffer pH 3.6, 10 mM TPTZ in 40 mM HCl and 20 mM $FeCl_3$ at a ratio of 10:1:1) were added to 0.1 ml of sample or standard. Absorbance was read at 593 nm for 10 min and the decay profile was fitted to a second-order phase model to obtain the value of absorbance at equilibrium. Ascorbic acid solutions ranging from 0.6 to 150 mg/ml were used to create a calibration curve. The results were expressed as milligrams of vitamin C equivalent antioxidant capacity (VCE) per 100 g HE ds.

For comparative purposes, both the measurements of bioactive compounds and the antioxidant capacity were expressed in terms of the HE dry solids in order to avoid the effect of "dilution by blend" [25]. Thus, the mass of formulated powders was corrected considering the fraction of HE.

2.5.1. Analysis of color properties

Color parameters were evaluated in dry (by reflectance) and in reconstituted (by transmittance) forms using an Evolution 600 UV-Vis Spectrophotometer (Thermo scientific, Waltham, USA). Solid samples were placed in glass plates with the same transparency. The color measurements were performed with a DRA-EV-600 Diffuse Reflectance Accessory (Thermo scientific, Waltham, USA) over the 220 - 850 nm spectrum at a 2° observation angle with a D65 illuminant. On the other hand, liquid samples were prepared by diluting the powder in doubly distilled water reaching the same Hibiscus solid extract (1 % w/v). Samples were hydrated, centrifuged, filtered through a 0.45 µm filter, and placed in a 1 cm path length glass cuvette. Color parameters of solids and liquids were obtained over the 300 - 800 nm spectrum with VISIONLite ColorCalc Software (ascanis OHG, Überlingen, Germany). The color parameters chroma (*C*), hue (*h*) and the total color difference ΔE were calculated according to Eqs. (3), (4), and (5):

$$C = \sqrt{(a^*)^2 + (b^*)^2} \quad (3)$$

$$h = tan^{-1}\left(\frac{b^*}{a^*}\right) \quad (4)$$

$$\Delta E = \sqrt{(L^* - L_0^*)^2 + (a^* - a_0^*)^2 + (b^* - b_0^*)^2} \quad (5)$$

Where L* (lightness), a* (redness/greenness) and b* (yellowness/blueness) are CIEL*a*b* coordinates. Subscript 0 indicates the control sample. For liquid/ reconstituted samples, the control was the freshly prepared extract (F-HE). For powders, the control was the extract with no carriers (HE).

2.6. Fourier-transform infrared spectroscopy

Fourier-transform infrared spectroscopy (FT-IR) measurements were performed in an FT-IR NICOLET iS5 (Thermo Scientific, Madison, USA). Individual components and obtained powders were studied by a single-bounce NICOLET iD3 ATR system (Thermo Scientific, Madison, USA) of ZnSe crystal and with an incident angle of 45 °. The spectra of samples were obtained by taking an average of 16 scans at a resolution of 4 $cm^{-1}$. The spectra were acquired between 600 and 4000 $cm^{-1}$. Spectral analysis was performed using the Thermo Scientific OMNIC software (Madison, WI, USA).

2.7. Storage stability of spray dried powders

Dehydrated samples were stored with and without exposure to light for 60 days at 25 ± 1 °C in a controlled temperature chamber. For storage under light, powders were spread in hermetically sealed glass petri dishes (55 mm x 10 mm) with the same transparency, size and headspace. A 12 W daylight LED lamp (OSRAM, China) was used, and the luminous flux' per unit area of 3500 ± 10 lx was determined by a digital light meter Cem dt-1301 (Shenzhen, China). For storage in darkness, samples were kept in the same chamber, in identical glass petri dishes but covered with aluminum foils. The stability of powder was assessed in terms of total monomeric anthocyanins and antioxidant capacity ($ABTS^{\cdot+}$). Complementarily, the pigment deterioration was evaluated by measuring the Browning index (BI) and the Polymeric color (PC) as described in [23].

2.8. Statistical analysis

Data were reported as mean ± standard deviation. Experiments and analytical measurements were carried out in triplicate. Measurements uncertainty was calculated based on the standard deviation of the mean with a coverage factor $k$=2. One-way ANOVA with Tukey post-test were performed to determine the significant differences among formulations. Stability studies involving light/darkness and sample formulation, and their interaction, as sources of variation, were analyzed by two-way ANOVA. The level of significance was defined at $P>0.05$ (interval of confidence of 95%). All statistical analysis and experimental data fitting were performed through GraphPad Version 4 (GraphPad, Software Inc., San Diego, CA, USA).

## 3. Result and discussion

### 3.1. Effect of wall material composition on physical properties

At studied conditions, drying yield varied between 60 and 70 %, and this was similar to previously reported for spray dried hibiscus extracts [4]. Table 2 shows the moisture and $a_w$ of recently obtained powders.

The spray dried extract (HE) without any drying additive ~~agents~~, presented the highest water content due to the highly hygroscopic low molecular weight compounds (sugars and organic acids) extracted by decoction from Hibiscus tissues [26]. Samples containing MD and gum showed lower water content ($P<0.05$) than the control. As previously reported, MD efficiently reduces the water affinity decreasing the moisture retention in powders [21]. Despite both gums did not exert significant effects on moisture respect to HEMD system, a slight difference between samples containing G or GA was observed. MDG2 showed slightly higher moisture than MDGA2, and this was related to the higher protein content of G (13.8 % ds.) compared to GA (1.37 % ds.) [2]. According to Karrar et al. [19], spray dried powders containing proteins show higher moisture than those containing only carbohydrates. This author related the high water holding capacity with the presence of proteins in an amorphous state. In addition, $a_w$ values were also different among the obtained powders and varied proportionally with moisture. In all cases, the $a_w$ values were lower than 0.3, indicating a good stability against microbial growth or physicochemical changes [5, 19].

As it is well known, the type of carrier and microstructure of particles determine most of the powders-solvent interaction properties and, consequently, their resultant functional properties [19]. In this sense, Table 2 shows the hygroscopicity, wettability, and solubility values determined for the examined samples. The composition of the powder affected significantly the hygroscopicity ($P<0.05$). HE presented the highest tendency to adsorb water ~~moisture~~ from the environment during storage for 7-day at 75 % RH. In the presence of MD, a significant decrease in water adsorption was noticed for HE, with no marked differences observed for formulations containing G or GA. The lower hygroscopicity, the lower the susceptibility to undergo undesirable physical changes (aggregation, caking problems, or stickiness) during shelf life [5], thus, powders with low hygroscopicity may have benefits for handling and storage.

Wettability measures the ease of particle penetration in water. It depends on several physical (size, density, porosity) and compositional (presence of hydrophilic groups, molecular interactions, moisture) properties [6]. As shown in Table 2, the obtained powders showed different wettability values. MD

reduced the time required for HE particles immersion in the solvent, while increasing G proportion resulted in longer times. Comparatively, MDG2 presented lower wettability than MDGA2. Despite this, observed wettability values were comparable to those reported by other spray dried Hibiscus formulations (285 – 765 s) [19].

Solubility is the last step in the particle dissolution process and it constitutes a decisive aspect in the functional quality assessment of powders. All the studied formulations showed good solubility (> 95 %) [19]. Although no marked differences were found for most of the studied systems, a slight increase in solubility was observed for those samples containing the highest amount of G (MDG3) respect to the dry extract (HE). This could indicate that G slightly improved the dispersion and dissolution capacity of the dehydrated extract.

Solvent-interaction properties could be interpreted considering the particle size distribution. In all cases, size distribution was monomodal and mesokurtic, except for MDGA2 (leptokurtic). In the absence of drying agents, HE showed the highest mean particle diameter (Table 2), while it was markedly reduced when MD was added. HEMD mean diameter was higher than values reported by Diaz-Bandera et al. [27] for Roselle-MD powders (7 μm) containing an extract-carrier ratio of 1:1, which is different from the relation used in this study (2.5:1).

The studied gums influenced the particle size. The higher proportion of G in the carrier mixture, the lower the particle diameter. As indicated by do Valle Calomeni, et al. [21], MD increases feed viscosity, resulting in larger average particles at high concentrations. Thus, increasing G with relative reduction of MD in the carrier mixtures may contribute to explain the lower size of particles observed. Comparatively, the composite formulations containing GA (MDGA2) resulted in markedly bigger particles than G (MDG2). As indicated by de Moura et al. [12], while larger-sized microparticles generally provide better protection than smaller ones, the smallest may result in better functionality. In this case, the lowest mean size of MDG3 could explain the improved solubility (Table 2).

Micrographs of powders obtained by spray drying are shown in Figure 1. In the absence of carrier agents, HE was observed as a continuous net with interparticle bridge formation. Cano-Chauca, Stringheta, Ramos, and Cal-Vidal [28] observed similar formations with large, amorphous, piled-up and strongly bounded particles at the exit of the spray dryer. These authors correlated those microstructural properties with high stickiness and cohesive forces among particles.

Both MD and gums allowed obtaining discrete, heterogeneous size and spherical-shaped particles. In fact, the sphericity factor obtained by morphological analysis was higher for formulated powders (0.66 - 0.68) than for HE (0.47). Consistent with our observations, Navidad-Murrieta et al. [6] also reported smooth surfaces when MD and GA were used as carrier agents. Similar to this study, particles showed concavities with wrinkles but without breaks or fissures. The corrugated appearance was previously attributed to shrinkage due to the fast water evaporation in the spray drying process, while wrinkles evidence the slow formation of a cover film when droplets are atomized [13]. Regardless of the presence, ratio, or type of gum, SEM evidenced a slight particle agglomeration in all formulations. Navidad-Murrieta et al. [6], who also studied the spray-drying of *Hibiscus sabdariffa* extracts with MD and GA, attributed this effect to the high outlet temperatures, which might cause adherence among particles, especially when they retained high moisture and have low glass transition temperatures

Collapse, agglomeration and stickiness, among other structural changes, occur when amorphous materials change from a very viscous glassy state to a rubbery (supercooled liquid) state due to being subjected to storage temperatures near o above the glass transition temperature ($T_g$). Thus, $T_g$ is an indicator of stability during long-term storage. It depends on moisture content, chemical structure and molecular weight of materials [19]. To explore the impact of powder composition on glass transition, $T_g$ was assessed by DSC in samples equilibrated at the same relative humidity (11 % RH) (Table 2).

The dried extract (HE) showed the lowest $T_g$ value, whilst MD increased it markedly ($P<0.05$). The presence of gums did not modify significantly the $T_g$ of the formulations containing MD, except for those samples with the highest proportion of G. The slight reduction in $T_g$ value in MDG3 could be attributed to the higher moisture retention, even at the same $a_w$ or RH. MD and GA have been commonly used to increase the $T_g$ of dehydrated natural extracts due to their high molecular weight [19]. In this study, no significant differences were found between G and GA, indicating that both gums similarly affect spray dried extract physical stability.

### 3.2. Bioactive compounds and antioxidant capacity

Table 3 shows the bioactive compounds and the antioxidant capacity of the obtained spray dried powders. Data from a freshly prepared aqueous extract (F-HE) was included for comparative purposes. In all cases, the results were expressed based on HE total dry solids, even in those formulations containing carrier agents as indicated in section 2.5.

In all cases, the spray dried samples showed an increased content of anthocyanins, polyphenols, and antioxidant capacity, regarding freshly prepared extract. This increase may be explained considering the thermally induced release of antioxidant compounds from insoluble forms (bounded to proteins, fibers or cell components) which remain in the decoction medium [29]. The exposure to hot air stream [3] along with the acidity of the medium (pH = 2.3 ± 0.1) may promote the hydrolysis and release of Hibiscus bioactive compounds [6].

Although direct comparisons between freshly prepared Hibiscus extract and spray dried powders were neglected or scarcely discussed in similar published works, interesting increases in bioactive compounds and/or antioxidant capacity were found. For instance, in Cid-Ortega et al. [4], Hibiscus ethanolic extract showed 5.6 mg cyanidin-3-O-glucoside /g ds., and 41 mg GAE/g ds., which increased after spray drying to 6.6 mg cyanidin-3-O-glucoside /g ds., and 49 mg GAE/g ds., respectively. Similarly, Ochoa-Velasco et al. [9] observed an increase in the antioxidant activity comparing hydro-alcoholic Hibiscus extract (28 mmol Trolox/100 g ds.) with spray dried powders (44 mmol Trolox/100 g ds.). These authors attributed these results to the generation of pores in powder during spray drying, which improved the extraction process and dissolution.

Among the obtained spray dried samples, the composite formulations showed a lower proportion of bioactive compounds and antioxidant capacity than HE, but no significant differences between the different carriers studied were observed (Table 3). Most of previous works also report a marked reduction in the phenolic or anthocyanin content as the proportion of carrier mixture increased [4, 9]. After ruling out the effect of dilution by blend, the bioactive reduction observed in this work was explained considering the thermally induced interactions among Hibiscus extract and excipients, which may hindered the extractability in aqueous media. Consistent with our observations, Diaz-Bandera et al. [27] studied the release mechanisms of Hibiscus polyphenols from spray dried powders with different carrier agents (proteins and polysaccharides) and reported that all carriers, even at maximum release equilibrium, retained polyphenols in different extent. Ochoa-Velasco et al. [9] justified similar observations in terms of powder microstructure. These authors proposed that high concentrations of the carrier (mesquite gum) could create a crystalline surface that decreases the extractability of bioactive compounds. Similarly, Navidad-Murrieta et al. [6] reported that carrier agents interact with phenolic compounds reducing the hydroxyl groups available to react with radicals, which decreases the antioxidant capacity of samples.

In this study, phenolic compounds ranged from 34 to 41 mg GAE/g HE ds. being these values slightly higher than those previously reported for spray dried Hibiscus sabdariffa composite powders, which varied between 7.2 to 9.8 mg/GAE g ds. [27], 7.1 to 32.1 mg/GAE g ds. [6] or 23 to 38 mg/GAE g ds. [4]. Similarly, total monomeric anthocyanin varying between 15 and 17 mg/g HE ds., were higher than previously reported values for formulated Hibiscus spray dried powders, which ranged between 3.3 to 5.4 mg/g powder ds. [4], or 3.0 to 3.4 mg/g ds. [9]. Differences found in this work could be attributed to the Hibiscus variety, particle size, type of solvent for extraction, ratio sample: solvent, time and method of extraction, as well as the spray drying conditions, and formulation, among others.

As expected, a significant correlation between bioactive compound content and antioxidant activity was observed. Correlation coefficients were slightly higher for phenolic compounds, measured with $ABTS^{+\cdot}$ ($r^2$=0.99) and FRAP ($r^2$=0.97), than for monomeric anthocyanins ($r^2$=0.96 and 0.93), which could indicate that the antioxidant properties of studied systems were governed principally by phenolic compounds, than by anthocyanins. As previously reported for Hibiscus extracts, the radical quenching capacity and the chelating activity have been attributed mainly to flavonoids (catechin and quercetin), phenolic acids (gallic and chlorogenic acids), tannins and anthocyanins [6]. Despite the phenolic content of G was reported higher (9.55 mg GAE/g gum ds.) than that of GA (1.76 mg GAE/g gum ds.) [2], the low proportion of gum in the powder formulation may result in an unnoticeable contribution to phenolic content as well as to the antioxidant properties.

### 3.3. Chromatic attributes in dry and reconstituted forms

In addition to antioxidant properties and related potential health-promoting benefits, rich in anthocyanin Hibiscus aqueous extracts also impart beautiful colorations, and color becomes a relevant quality parameter [30]. Chromatic properties were assessed both in powdered and in reconstituted form (Table 3). Both, powder compositions and the physical state (solid or aqueous), modified the chromatic properties of the studied systems ($P<0.05$). Dissolution of samples accounted for 91 % of the total variance, indicating that dispersion in water induced the deepest changes in color attributes (Figure 2). Reconstituted samples showed lower luminosity but higher redness (a*), yellowness (b*), hue, and chroma than powders. Ochoa-Velasco et al. [9] who evaluated the chromatic attributes of spray dried and reconstituted Hibiscus extracts containing mesquite gum as a carrier agent reported similar changes.

In dehydrated systems, HE showed the lowest L* value in agreement with the visually perceived dark color (Figure 2). Higher lightness in formulated samples was attributed to the contribution of whitish maltodextrin as reported by Cid-Ortega et al. [4]. MDGA2 showed a further increase in L* value. The highest luminosity in samples containing GA concerning those containing G, was attributed to the fact that gum arabic is clearer (L*=94.3) than purified G (L*=83.6) [2]. Because of its dark color, G minimized the impact on color change of deep colored samples [9]. Formulated powders showed a higher a* than HE, and this was more noticeable for samples containing GA. Regardless subtle changes in b*, all samples kept in the red-yellow range (0 to 90°) of hue angle. The increase in chroma, indicating brighter and pure colors, could be related with the increase in luminosity of the samples, and could imply a positive attribute for a food powder [12]. Among dehydrated samples, MDGA2 showed the highest total color difference (ΔE) from spray dried HE.

In reconstituted systems, the color changes according to the formulation followed a similar trend to that observed in dry. The chromatic attributes of aqueous samples were assessed also in freshly prepared Hibiscus aqueous extract (F-HE). F-HE evaluated at the same total solid HE concentration, showed a brighter red color, which was visually different to reconstituted ones. Reconstituted samples showed lower L* but higher a* and b* values. In agreement, changes in hue also indicates a shift toward red values, which could be attributed to the thermal release of anthocyanins mentioned earlier, as well as to pigmented derivatives from dried carriers that modify de color.

### 3.4. Fourier-transform infrared spectroscopy

The chemical structure of individual components (MD, G and GA), the spray dried Hibiscus extract (HE), and formulated powders (HEMD, MDG2 and MDGA2) were examined by FT-IR. The infrared spectra are shown in Figure 3.

MD spectrum showed typical bands of polysaccharides at 3270 $cm^{-1}$ (O-H stretching), 2900 $cm^{-1}$ (carboxylic C-H stretching), 1632 $cm^{-1}$ (C=O stretching), 1350 $cm^{-1}$ (O-H bending) and 1150 $cm^{-1}$, 1078 $cm^{-1}$ and 990 $cm^{-1}$ attributable to C-O stretching and C-O-H bending in agreement with Mahdi et al. [5]. As previously reported, the G and GA spectrums were similar, evidencing a common chemical nature [1]. Gums showed a wide band, centered at 3300 $cm^{-1}$ (–OH stretching) and a band at 2930 $cm^{-1}$ (-CH stretching) which were sharper in G. The bands at 1600 $cm^{-1}$ and 1400 $cm^{-1}$ were attributed to -C=O

asymmetrical and symmetrical stretching, respectively. Finally, the peak observed at 980 $cm^{-1}$ in G and 1015 $cm^{-1}$ in GA was attributed to O-H bending [19].

HE showed a wide peak between 2900 and 3600 $cm^{-1}$ attributable to bonded -OH and typical bands at 2932, 1403, and 1059 $cm^{-1}$ due to –CH, C-C and C-O groups, respectively [31]. According to Fragoso et al. [32], bands observed between 1175 and 1400 $cm^{-1}$ may correspond to phenolic compounds such as anthocyanins and condensed tannins. Particularly, the bands at 1606 and 1185 $cm^{-1}$ may be assigned to benzene ring, and C=C bonds of aromatic rings [33]. Finally, bands in the region between 1300 and 1460 $cm^{-1}$ can be imputed to the presence of hydroxycinnamic acids [12].

Spray dried powders showed characteristic bands of conformational structure of MD, G and GA (Figure 3A), as indicated by points 1, 2, 3 and 4 in Figure 3A. Typical bands of HE were also noticed in formulated powders (points 5, 6, 7 and 8 in Figure 3B), indicating that the extract was dispersed both in the polymeric matrix and at the particles surface. Despite Hibiscus extract was the main component in all spray dried powders, profiles of HEMD, MDG2 and MDGA2 were quite different regarding HE. Band displacements (point 9) and changes in the intensity of peak (point 10) in the spectrum of powders compared to HE were observed. Most of these changes occurred in the range of 800 to 1150 $cm^{-1}$, corresponding to characteristic polysaccharides groups (C-O, C-O-C) [26], as well as in the range of 1130 to 1460 $cm^{-1}$, related to phenolic compounds [32]. Present results confirmed that molecular interactions between Hibiscus extract compounds and carrier agents occurred.

### 3.5. Storage stability of Hibiscus powders

Figure 4 summarizes the changes in Total monomeric anthocyanins (Figure 4A), $ABTS^{\cdot+}$ scavenging activity (Figure 4B), Polymeric color (Figure 4C) and Browning Index (Figure 4D) undergone in samples after 60 days of storage both in dark and light conditions at 25 °C. Initial values are depicted with black bars, while values for stored samples in darkness or light are depicted with dark or light gray, respectively.

As shown in Figure 4A, the monomeric anthocyanins decreased with time for all systems. Light effect and sample composition accounted for 44 and 42 % of the variation, indicating a significant impact on the pigment loss ($P<0.001$). As expected, the samples stored in darkness (dark grey bars) exhibited higher pigment protection [30]. In the darkness, the studied systems showed pigment losses from 5 to 6 %, except for those containing G, which did not show significant differences. Light exposure intensified the

losses of anthocyanins in all systems (light grey bars). This effect was more noticeable for HE and MDGA (11 %) than for HEMD and samples containing G, for which pigment loss was about 8 %. As shown, G contributed to a relative improvement in anthocyanin retention, and this was attributed to the complex mixture of non-anthocyanin polyphenols present in the gum [2]. Phenolic compounds in *P. alba* gum, despite their low proportion, could improve the anthocyanins' stability by acting as co-pigments or antioxidants. As reported, phenolic compounds form non-covalent complexes with the anthocyanins by vertical stacking of π-electron in the feed solution and survive the spray drying process [30]. Copigment-anthocyanins complexes prevent the hydration of the flavilium moiety contributing to stabilizing the pigment. Similarly, Weber et al. [30] investigating the potential of phenolic compounds as co-pigments, observed an enhanced storage stability of a spray dried anthocyanin powder obtained from a blackberry extract. In addition, phenolic compounds may also contribute to pigment stability by reducing the oxidative damage of anthocyanins. Thus, the antioxidant compounds in G may report an additional positive effect when this gum is used as a carrier agent.

Figure 4B shows how antioxidant activity of powders, measured in terms of ABTS scavenging activity, also decreased significantly after storage. This reduction was mainly influenced by powder composition (72 % variation) than light exposure (3 % variation). Similar results were reported by Jiménez-Aguilar et al. [25] who studied the impact of storage on spray dried blueberry extract at 25 °C with/without light. The highest losses of antioxidant capacity was observed for HEMD (16 %) and MDGA2 (14 %), followed by HE (8 %). Samples containing G showed lower average losses, 5 % and 7 % for dark and light storage, respectively, which could indicate that G contribute to preserve the antioxidant capacity of the obtained powders.

Figure 4C depicts that Polymeric Color index (PC) changed in powders both by composition as well as by light exposure ($P<0.001$). After storage in darkness, PC increased significantly for all samples except for MDG1 and MDG2, which could indicate that low and intermediate concentrations of G prevented the formation of polymeric-colored substances. Light exposure, augmented significantly the PC in all samples, and this was more noticeable for samples containing G. PC was related to condensation reactions of anthocyanins themselves, as well as with other phenolic compounds (flavan-3-ols, polyflavan-3-ols, tannin, phenolic acids, etc.), or with non-phenolic (amino acids and proteins) compounds [34]. The formation of colored derivatives involving anthocyanin degradation agreed with the lowest retention of anthocyanin monomers observed in HE and MDGA2. In the case of samples

containing G, the light-induced PC increase seems not to be strictly related to anthocyanin degradation. Hence, further studies are needed to gain insight into the polymeric color increase mechanism in samples containing G.

Finally, the Browning Index (BI), as indicative of pigment stability was analyzed and is shown in Figure 4D. In all samples, the BI increased after 60 days of storage, indicating that browning of powders occurred [11]. Both, powder composition and light exposure, had a significant effect ($P<0.001$), accounting for 31 % and 62 % of the total variance, respectively. HE showed the highest starting BI value, but after storage, the increase of BI in darkness (27 %) and light (47 %), were comparatively lower than for formulated samples. HEMD showed a sharp BI increase regardless of light/dark storage. In samples containing G, the BI increased by 37, 50 and 60 % as the G proportion increased, and this trend was more evident by light exposure. MDGA2 showed similar BI to MDG2, both for dark and light storage. The formation of brown color was related to the loss of anthocyanin monomers, which may involve copigmentation of anthocyanins with other phenolic compounds [11].

## 4. Conclusions and future directions

*Prosopis alba* exudate gum and maltodextrin enabled to obtain low moisture, low water activity and highly soluble rich-in-antioxidant pigment powders by spray drying of Hibiscus aqueous extract. Particles containing high content of polyphenols and anthocyanins, showed better stability of antioxidant properties in presence of G, even for 60 days of storage exposed to light and 25 °C. With these results, it is of interest to study formulations with higher proportion of G and conclude about its properties and bioavailability. Results of present work encourage considering G as a feasible component of carrier blends for spray drying rich-in-antioxidant aqueous pigments extracts. Such insights could be harnessed to design functional ingredients with specific characteristics for sought applications in food, pharmaceuticals, cosmetics, among other industries. The use of *P. alba* exudate gum at an industrial scale would also have positive effects on local economies, not only by providing an alternative to currently imported additives, but by adding value to a currently wasted or minimally used resource.

**ACKNOWLEDGMENTS**

The authors acknowledge the financial support of Universidad Nacional del Chaco Austral (UNCAUS – PI N°79/18 and 161/22), Universidad de Buenos Aires (UBACYT N° 20020170100557BA), Agencia

Nacional de Promoción Científica y Tecnológica (PICT 2020 N°01909, 2017 N°1744), Consejo Nacional de Investigaciones Científicas y Técnicas (PIP 11220200101351CO), Thematic Network CYTED (4150495), and IPN-SIP (20181309).

**Conflict of Interest.** The authors declare no competing interests.

**Data Availability Statement.** The data supporting the findings of this study are available on request from the corresponding author.

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**FIGURE CAPTIONS**

**Fig. 1.** Scanning electron microscopy (magnification x500, x1k, x2.5k) of spray dried powders of Hibiscus extract (HE), Hibiscus extract containing maltodextrin (HEMD), maltodextrin and *Prosopis alba* exudate gum (MDG2) or maltodextrin and gum arabic (MDGA2).

**Fig. 2.** Color cards illustrating the color of powders in dry or reconstituted form of Hibiscus extract (HE), Hibiscus extract containing maltodextrin (HEMD), maltodextrin and *Prosopis alba* exudate gum (MDG) or maltodextrin and gum arabic (MDGA). F-HE is the color of fresh extract. Colors displayed illustrate the chromatic coordinates according to a free color converter software (https://www.nixsensor.com/free-color-converter/).

**Fig. 3.** FT-IR spectra of individual maltodextrin (MD), *Prosopis alba* exudate gum (G), gum arabic (GA), Hibiscus extract (HE), and formulated powders Hibiscus extract containing maltodextrin (HEMD), maltodextrin and *Prosopis alba* exudate gum (MDG) or maltodextrin and gum arabic (MDGA).

**Fig. 4.** Total monomeric anthocyanin (A), $ABTS^{\cdot+}$ radical scavenging activity (B), Polymeric color (C), and Browning Index (D) of spray dried powders of Hibiscus extract (HE), Hibiscus extract containing maltodextrin (HEMD), maltodextrin and *Prosopis alba* exudate gum (MDG) or maltodextrin and gum arabic (MDGA). Before (■) and after 60 days of storage in dark (■) or under light (■). The error bars represent the measure uncertainty.

**Table 1**

Most recent studies related to obtaining functional ingredients rich in antioxidants and pigments from *Hibiscus sabdariffa* extracts (core) through drying or ionic gelation. AS: Achira starch, CA: carrageenan, CMC: carboxymethyl cellulose, GA: gum arabic, GE: gelatin, MD: maltodextrin, MG: mesquite gum, PE: pectin, RO: rapseed oil, WP: whey protein.

| Encapsulation method | Carrier agents | Studies on physicochemical and structural properties | Studies on phenolic compounds and antioxidant properties | Stability studies | Ref. |
|---|---|---|---|---|---|
| Spray-drying | MD, PE, GE, CMC, WP, CA, GA | ✓ | ✓ | × | [27] |
| Spray-drying | MD, GA | ✓ | ✓ | × | [4] |
| Spray-drying | MD, GA | ✓ | ✓ | × | [6] |
| Spray-drying | MG | ✓ | ✓ | ✓ | [9] |
| Spray-drying | AS | ✓ | ✓ | × | [13] |
| Ionic gelation | RO, PE, | ✓ | ✓ | ✓ | [12], [14] |

✓: Included, ×: Not Included.

**Table 2**

Physicochemical properties of spray dried Hibiscus extract (HE), and formulated powders containing HE and different proportions of maltodextrin (MD), *Prosopis alba* exudate gum (G), or arabic gum (GA): 100% MD (HEMD), 95% MD 5% G (MDG1), 90% MD 10% G (MDG2), 85% MD 15% G (MDG3) and 90% MD 10% GA (MDGA2).

| | **HE** | **HEMD** | **MDG1** | **MDG2** | **MDG3** | **MDGA2** |
|---|---|---|---|---|---|---|
| *Moisture, % ds.* | $5.88 \pm 0.53^{a}$ | $4.46 \pm 0.16^{b,c}$ | $4.64 \pm 0.11^{b,c}$ | $4.90 \pm 0.22^{b}$ | $5.00 \pm 0.39^{b}$ | $4.10 \pm 0.22^{c}$ |
| $a_w$ | $0.25 \pm 0.00^{b,c}$ | $0.24 \pm 0.00^{d}$ | $0.24 \pm 0.00^{c,d}$ | $0.26 \pm 0.00^{a,b}$ | $0.27 \pm 0.00^{a}$ | $0.22 \pm 0.01^{e}$ |
| Hygroscopicity, *% ds.* | $26.5 \pm 0.02^{a}$ | $20.8 \pm 0.68^{c}$ | $20.7 \pm 0.35^{c}$ | $21.0 \pm 0.04^{b,c}$ | $21.5 \pm 0.53^{b,c}$ | $21.9 \pm 0.29^{b}$ |
| Wettability, *s* | $482 \pm 28^{d}$ | $276 \pm 68^{c}$ | $573 \pm 11^{b,c}$ | $745 \pm 90^{a,b}$ | $894 \pm 90^{a}$ | $274 \pm 10^{c}$ |
| Solubility % ds. | $95.2 \pm 1.11^{c}$ | $96.8 \pm 0.60^{d}$ | $95.3 \pm 0.25^{c}$ | $96.6 \pm 1.31^{b}$ | $97.8 \pm 0.08^{a}$ | $95.8 \pm 0.70^{d}$ |
| Mean diameter, µm | 96.1 ± 33.2 | 26.9 ± 13.7 | 34.4 ± 19.3 | 24.8 ± 12.9 | 17.[illegible] ± 7.97 | 54.7 ± 20.0 |
| Sphericity factor | 0.47 | 0.66 | 0.66 | 0.67 | 0.68 | 0.66 |
| $T_g$ (°C), *11% RH* | $46.4 \pm 0.71^{a}$ | $53.1 \pm 0.98^{b}$ | $52.9 \pm 0.16^{b,c}$ | $52.1 \pm 0.2[illegible]^{b,c}$ | $50.8 \pm 0.31^{c}$ | $52.6 \pm 0.10^{b,c}$ |

Mean ± SD values followed by different letters within the same row are significantly different according to ANOVA at $P \leq 0.05$. Maximum experimental uncertainty is estimated with ± 0.01 in the measuring conditions.

**Table 3**

Bioactive compounds, antioxidant capacity and color attributes of freshly prepared extract (F-HE), spray dried Hibiscus extract (HE), and formulated powders containing HE and maltodextrin (MD), *Prosopis alba* exudate gum (G) or arabic gum (GA), at different proportions: 100% MD (HEMD), 95%MD 5%G (MDG1) 90%MD 10%G (MDG2), 85%MD 15%G (MDG3) and 90%MD 10%GA (MDGA2). ΔEp: Total color difference for powder samples was calculated taking HE as reference. ΔEs: Total color difference for reconstituted samples was calculated taking F-HE as reference.

| | F-HE | HE | HEMD | MDG1 | MDG2 | MDG3 | MDGA2 |
|---|---|---|---|---|---|---|---|
| Total polyphenols, mg GAE/g HE ds. | 22.2 ± 1.2$^{c}$ | 40.6 ± 1.1$^{a}$ | 36.5 ± 1.4$^{b}$ | 34.4 ± 0.3$^{b}$ | 35.3 ± 0.7$^{b}$ | 35.9 ± 1.2$^{b}$ | 35.2 ± 0.7$^{b}$ |
| Total anthocyanins. mg cyanidin-3-O-glucoside /g HE ds. | 2.2 ± 0.0$^{c}$ | 17.3 ± 0.2$^{a}$ | 15.5 ± 0.1$^{b}$ | 15.1 ± 0.0$^{b}$ | 15.3 ± 0.0$^{b}$ | 15.4 ± 0.2$^{b}$ | 15.6 ± 0.2$^{b}$ |
| TEAC, mmol TE/ 100 g HE ds. | 15.5 ± 0.7$^{c}$ | 28.0 ± 0.1$^{a}$ | 25.3 ± 1.1$^{b}$ | 24.1 ± 0.6$^{b}$ | 24.3 ± 0.3$^{b}$ | 23.7 ± 0.7$^{b}$ | 24.5 ± 1.0$^{b}$ |
| FRAP, mg VCE/ 100 g HE ds. | 22.3 ± 1.2$^{c}$ | 44.0 ± 0.8$^{a}$ | 34.6 ± 1.9$^{b}$ | 35.4 ± 2.4$^{b}$ | 35.7 ± 1.0$^{b}$ | 35.4 ± 1.2$^{b}$ | 36.5 ± 0.3$^{b}$ |
| Color Powder | | | | | | | |
| L* | - | 53.6 ± 0.2$^{c}$ | 57.3 ± 0.1$^{b}$ | 57.3 ± 0.1$^{b}$ | 57.3 ± 0.1$^{b}$ | 57.4 ± 0.1$^{b}$ | 58.1 ± 0.1$^{a}$ |
| a* | - | 25.2 ± 0.3$^{d}$ | 30.8 ± 0.2$^{b}$ | 30.5 ± 0.1$^{b,c}$ | 30.5 ± 0.1$^{b,c}$ | 30.3 ± 0.2$^{c}$ | 31.4 ± 0.1$^{a}$ |
| b* | - | 10.8 ± 0.1$^{d}$ | 13.0 ± 0.1$^{b}$ | 12.8 ± 0.0$^{b,c}$ | 12.7 ± 0.0$^{c}$ | 12.7 ± 0.0$^{c}$ | 13.2 ± 0.1$^{a}$ |
| Hue | - | 23.2 ± 0.1$^{a}$ | 22.8 ± 0.1$^{b,c}$ | 22.8 ± 0.1$^{b}$ | 22.6 ± 0.1$^{d}$ | 22.7 ± 0.1$^{c,d}$ | 22.8 ± 0.1$^{b,c}$ |
| Chroma | - | 27.4 ± 0.4$^{d}$ | 33.4 ± 0.1$^{b}$ | 33.0 ± 0.1$^{b,c}$ | 33.0 ± 0.1$^{b,c}$ | 32.9 ± 0.2$^{c}$ | 34.1 ± 0.1$^{a}$ |
| ΔEp | - | - | 7.1 ± 0.2$^{b}$ | 6.8 ± 0.1$^{c}$ | 6.7 ± 0.1$^{c}$ | 6.6 ± 0.1$^{c}$ | 8.1 ± 0.1$^{a}$ |
| Color Solution (1 % HE w/v) | | | | | | | |
| L* | 45.6 ± 0.1$^{a}$ | 38.6 ± 0.1$^{f}$ | 40.2 ± 0.2$^{e}$ | 41.0 ± 0.1$^{c,d}$ | 41.2 ± 0.1$^{c}$ | 40.7 ± 0.1$^{d}$ | 42.0 ± 0.0$^{f}$ |
| a* | 53.4 ± 0.0$^{f}$ | 65.6 ± 0.1$^{e}$ | 66.2 ± 0.0$^{bc}$ | 66.3± 0.0$^{b}$ | 66.1 ± 0.0$^{c}$ | 65.7 ± 0.0$^{d}$ | 66.5 ± 0.1$^{a}$ |
| b* | 51.1 ± 0.1$^{e}$ | 66.1 ± 0.3$^{d}$ | 68.4 ± 0.4$^{c}$ | 69.8 ± 0.1$^{b}$ | 69.6 ± 0.4$^{b}$ | 69.3 ± 0.1$^{b,c}$ | 71.3 ± 0.1$^{a}$ |
| Hue | 43.7 ± 0.0$^{e}$ | 45.2 ± 0.1$^{d}$ | 45.9 ± 0.2$^{c}$ | 46.5 ± 0.2$^{b}$ | 46.5 ± 0.0$^{b}$ | 47.0 ± 0.1$^{b}$ | 46.1 ± 0.1$^{a}$ |
| Chroma | 73.9 ± 0.1$^{f}$ | 97.1 ± 0.3$^{e}$ | 95.2 ± 0.3$^{d}$ | 96.3 ± 0.1$^{b}$ | 96 ± 0.3$^{b,c}$ | 95.5 ± 0.1$^{c,d}$ | 97.4 ± 0.0$^{a}$ |
| ΔEs | - | 20.0 ± 0.2$^{d}$ | 22.2 ± 0.3$^{c}$ | 23.2 ± 0.1$^{b}$ | 22.9 ± 0.4$^{b,c}$ | 22.5 ± 0.0$^{b,c}$ | 24.3 ± 0.0$^{a}$ |

Mean ± SD values followed by different letters within the same row are significantly different according to ANOVA at P≤0.05. Maximum experimental uncertainty is estimated with ± 0.01 in the measuring conditions.

**Declaration of interests**

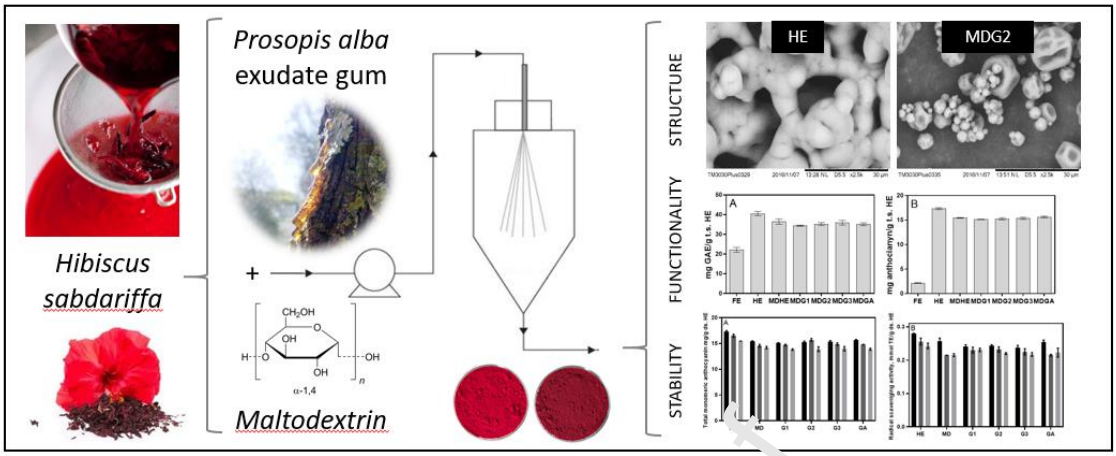
☒The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

**AUTHOR CONTRIBUTIONS**

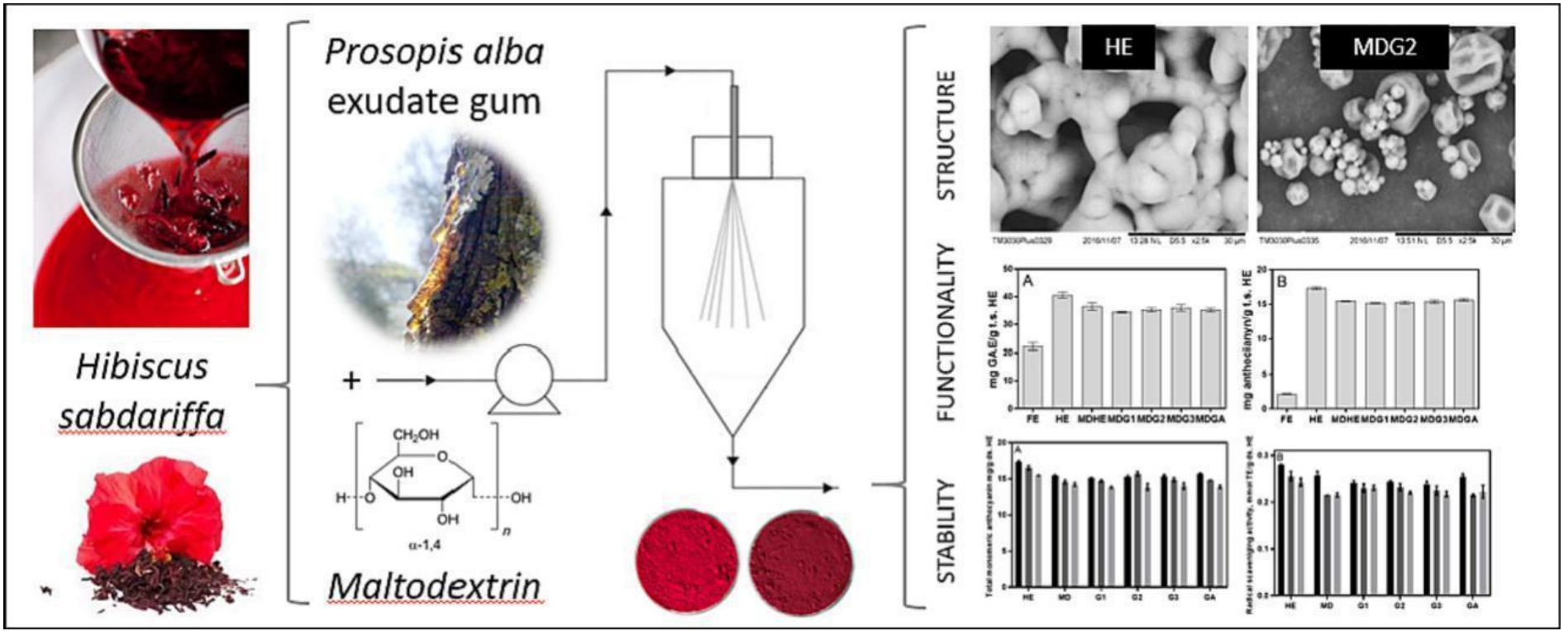
**Franco Emanuel Vasile**: Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original Draft, Funding acquisition **Diego Alberto Archaina**: Investigation **Jaime Jiménez-Guzmán**: Investigation **Gustavo Fidel Gutiérrez-López**: Resources, **Liliana Alamilla-Beltrán**: Conceptualization, Methodology, Resources, Supervision, Project administration, Funding acquisition **María Florencia Mazzobre**: Conceptualization, Resources, Writing - Review & Editing, Funding acquisition

## GRAPHICAL ABSTRACT

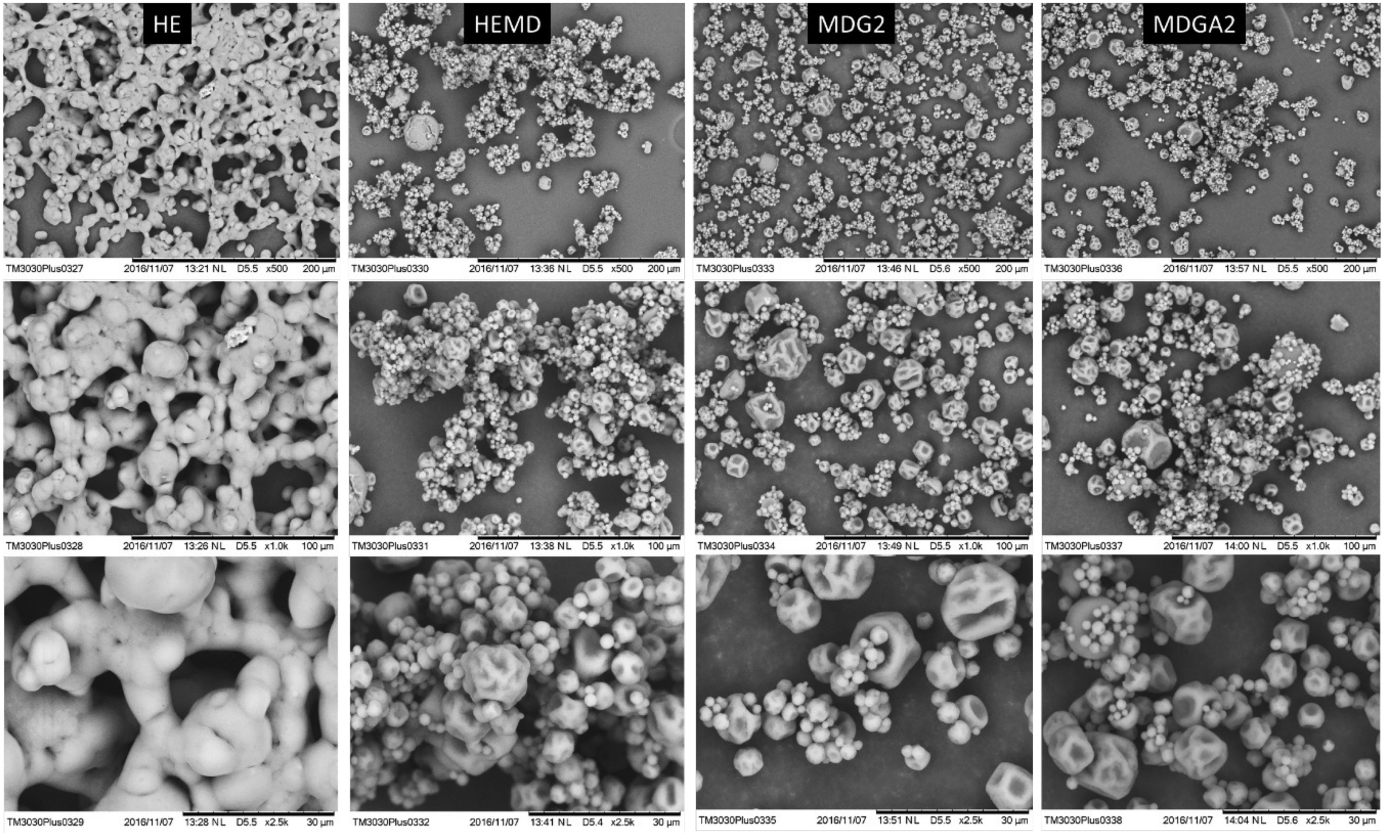


**HIGHLIGHTS**

- *Prosopis alba* exudate gum was used for spray drying *Hibiscus sabdariffa* extracts
- The studied gum affects the physicochemical and structural properties of powders
- *Prosopis alba* exudate gum behaves as carrier agent similar to arabic gum
- The studied gum preserves the chromatic and antioxidant characteristics of extracts




Graphics Abstract


HE
HEMD
MDG2
MDGA2
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TM3030Plus0333 2016/11/07 13:46 NL D5.6 x500 200 µm
TM3030Plus0336 2016/11/07 13:57 NL D5.5 x500 200 µm
TM3030Plus0328 2016/11/07 13:26 NL D5.5 x1.0k 100 µm
TM3030Plus0331 2016/11/07 13:38 NL D5.5 x1.0k 100 µm
TM3030Plus0334 2016/11/07 13:49 NL D5.5 x1.0k 100 µm
TM3030Plus0337 2016/11/07 14:00 NL D5.5 x1.0k 100 µm
TM3030Plus0329 2016/11/07 13:28 NL D5.5 x2.5k 30 µm
TM3030Plus0332 2016/11/07 13:41 NL D5.4 x2.5k 30 µm
TM3030Plus0335 2016/11/07 13:51 NL D5.5 x2.5k 30 µm
TM3030Plus0338 2016/11/07 14:04 NL D5.6 x2.5k 30 µm

Figure 1

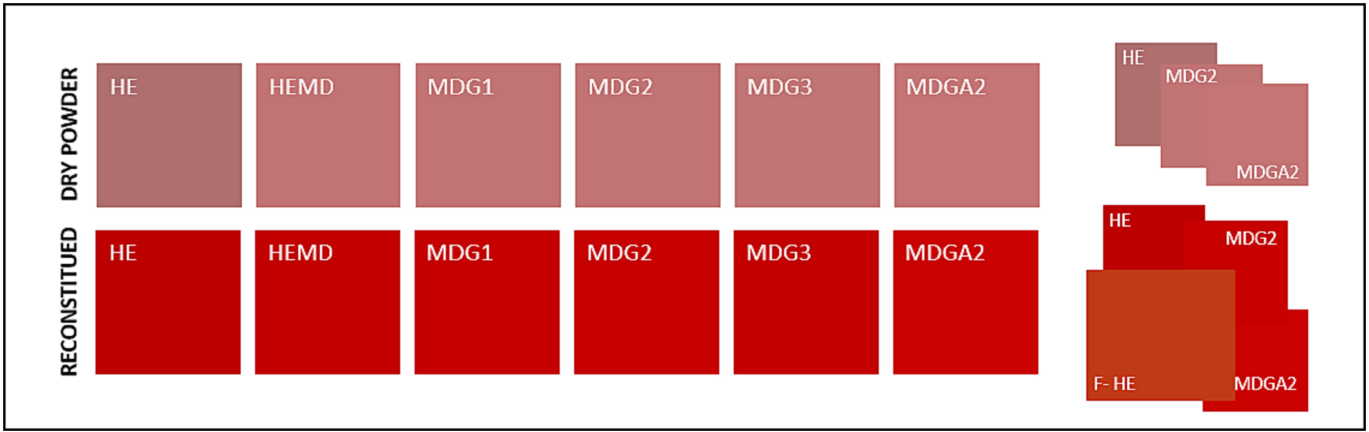
DRY POWDER
HE
HEMD
MDG1
MDG2
MDG3
MDGA2
RECONSTITUED
HE
HEMD
MDG1
MDG2
MDG3
MDGA2
HE
MDG2
MDGA2
HE
MDG2
F- HE
MDGA2

Figure 2

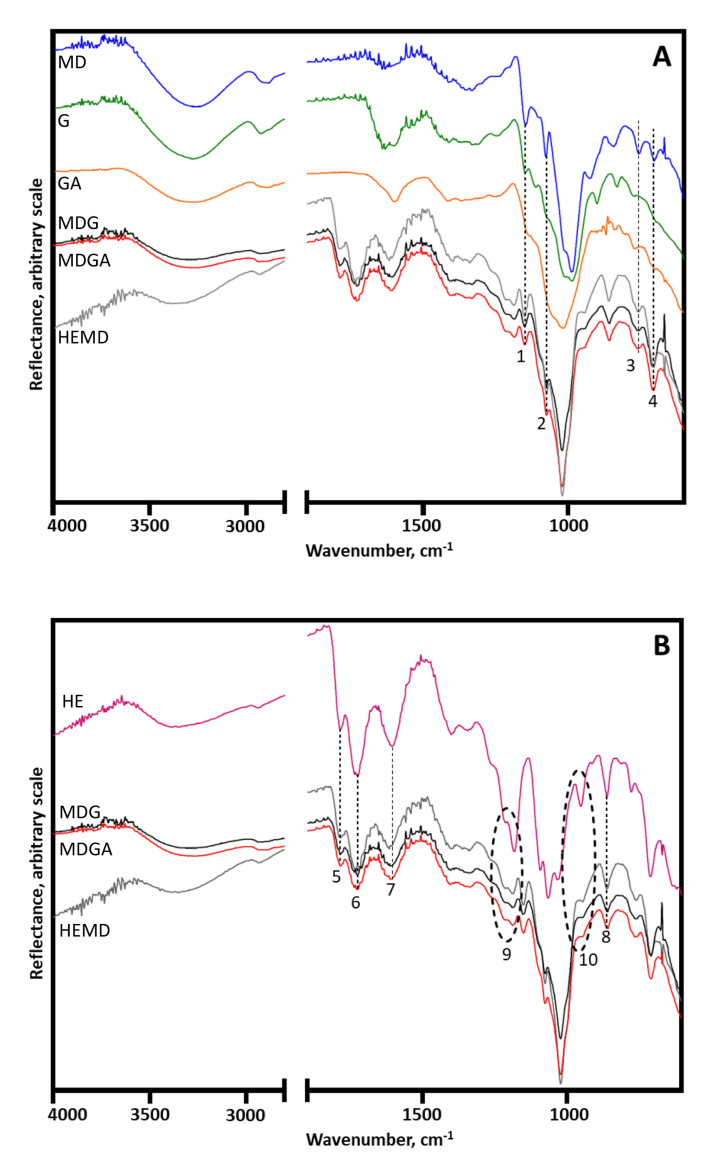
A
MD
G
GA
MDG
MDGA
HEMD
Reflectance, arbitrary scale
1
2
3
4
4000
3500
3000
1500
1000
Wavenumber, cm-1
B
HE
MDG
MDGA
HEMD
Reflectance, arbitrary scale
5
6
7
8
9
10
4000
3500
3000
1500
1000
Wavenumber, cm-1

Figure 3

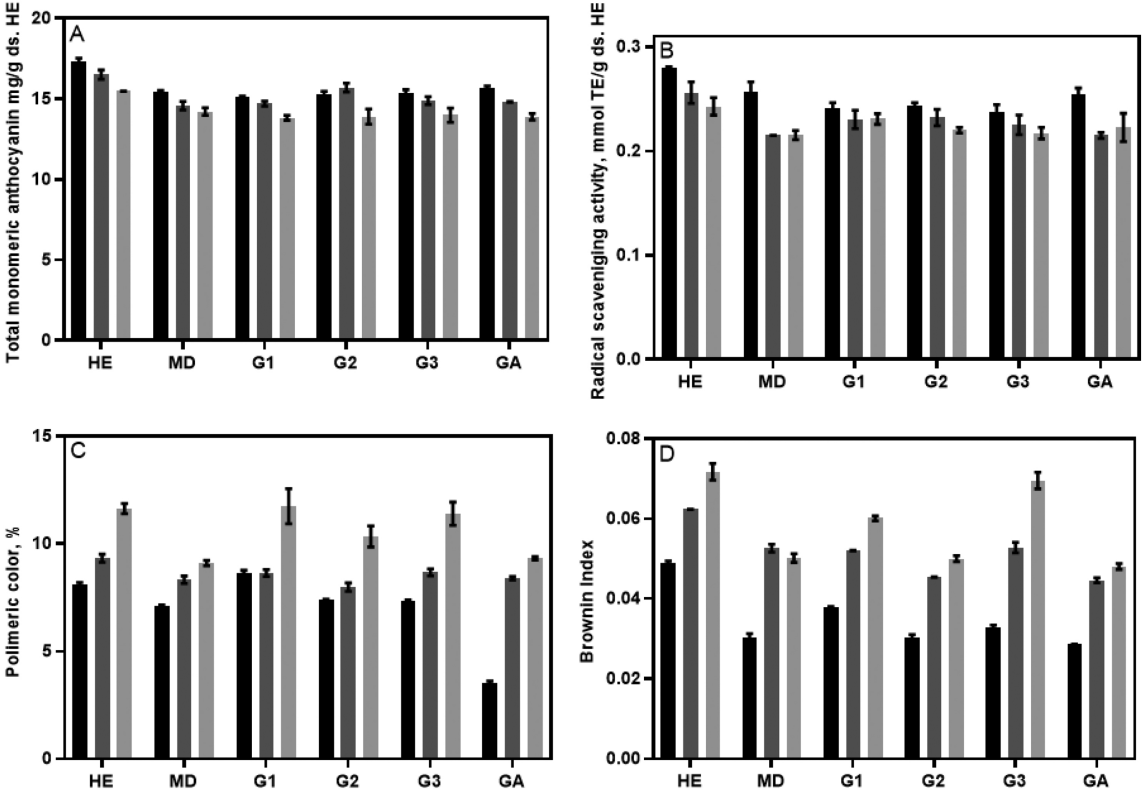
A
Total monomeric anthocyanin mg/g ds. HE
20
15
10
5
0
HE
MD
G1
G2
G3
GA
B
Radical scavenging activity, mmol TE/g ds. HE
0.3
0.2
0.1
0.0
C
Polimeric color, %
D
Brownin Index
0.08
0.06
0.04
0.02
0.00

Figure 4